

# MERCURY IN TUNAS: A REVIEW

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## ABSTRACT

Mercury in the aquatic environment comes from both natural processes and industrial activities. The latter probably have not significantly altered the mercury content of the high seas where most tunas are captured.

Mercury compounds enter aquatic organisms through the food web and/or by direct extraction from solution. The relative importance of these pathways in tunas is unknown. Mercury occurs in tuna principally in the form of methylmercury.

Generally, tunas appear to have higher mercury levels than those fish species which occupy a lower level in the food chain.

Mercury content of tunas varies according to fish size. However, other factors such as area of capture, differential growth rates, varying analytical techniques, and different sampling methods may account for some of the observed variation.

The U.S. Food and Drug Administration has established an "in-house" standard of 0.5 ppm of mercury for fishery products sold in the United States. Other countries have established limits as high as 1.0 ppm.

Seawater contains a wide array of dissolved elements and salts including mercury. Some of these occur in quantities that can be recovered on a commercial scale whereas the minute concentrations of others can be detected only by highly sophisticated techniques. Mercury does not play a prominent role among the metals dissolved in seawater because it occurs there only in very small quantities. Mercury in its various forms is also present in the earth's crust from which it can be dissolved by water. In the absence of water, especially under conditions of increased heat, mercury will enter the atmosphere in the form of vapor, from which it can dissolve in water at the water-air interfaces, i.e., rain or the sea surface. It is difficult to conceive that life could have evolved on this planet in an environment devoid of this ubiquitous element so it is of no great surprise that mercury is present in the bodies of living organisms. Furthermore, as is the case with many elements and their compounds, living organisms are even capable of concentrating mercury within or outside of their bodies.

Tunas are no exception—mercury occurs in various parts of these pelagic species in concentrations substantially exceeding those dissolved in the waters of the world oceans.

An epidemic of deaths and serious physical impairment among Japanese during the 1950's and 1960's (which was later related to the ingestion of seafood contaminated by mercury from local industries), the discovery of high levels of mercury in freshwater fishes and wildlife in Sweden in the 1960's, and the discovery of unusual mercury concentrations in fishes from parts of the Great Lakes system in 1970 led investigators to examine mercury in tunas and billfishes. The findings resulted in the seizure of some tunas and swordfish in the United States whose mercury content exceeded government standards, and significantly affected fishing strategy and marketing of these species.

## SOURCES OF MERCURY IN THE MARINE ENVIRONMENT

Mercury is most often encountered in nature as mercury sulfide or cinnabar. Other compounds and metallic mercury contribute a small percentage of the total available mercury.

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A number of natural processes release a continuous but relatively minor amount of mercury into the soil, groundwater, and air around deposits of mercury compounds. This effusion and volatilization of mercury is the first of many steps in the generation of its compounds and their subsequent entry into bodies of water including the oceans. This mercury is oxidized or recompounded in the environment, deposited in the sediments if insoluble or dispersed in the water if soluble. Rainfall causes erosion of soils and facilitates the mobilization of the mercury into the aquatic environment.

The second (and controllable) source of mercury in rivers, lakes, and oceans stems from the activities of man. Mercury compounds have been used for millennia as a coloring agent, as medicine, and for a multitude of industrial uses. The most common industrial sources of mercury contamination can be divided into several classes. Fossil fuels, i.e., coal and oil, have relatively low levels of mercury, but as millions of tons of these fuels are used each year, they release a substantial amount of mercury into the atmosphere which eventually enters the aquatic environment. Factories producing plastics, chlorine, caustic soda, and/or caustic potash have been charged with one of the more obvious and largest discharges of mercury and its compounds into rivers, lakes, and oceans. The fungicidal properties of mercury compounds have made it useful in preservation of paint and as a coating for seeds used in planting of crops. Mercury compounds also have slimicide properties which have brought about their extensive use in pulp mills and other industries. Gold, silver, and rare metals can be and are extracted by using metallic mercury in an amalgamation process in which the final step is driving off the mercury by heat to leave the other metals behind. The subsequent condensation of the mercury vapors is not 100% effective, and thus some of it enters the environment. Mercury contamination also results from many other industrial processes. However daily household, laboratory, and small business activities contribute considerable quantities of mercury and its compounds to sewage and sludge stores

by the breakage of thermometers and electric switches, discarding of or excretion of mercurials used for medication, and other seemingly unimportant events.

Studies show that the contaminant mercury is usually concentrated near the sites of outfall and that the levels decrease with distance from the sources.

## MERCURY IN FISHES

Methylmercury is one of the mercury compounds most often found in the bodies of fishes and other aquatic organisms (Westöo, 1966; Johnels et al., 1967). This compound is known to be readily derived from inorganic or metallic forms usually by biological intermediaries (Jernelöv, 1972a). Ionic mercury binds readily to organic materials and can be converted to methylmercury through bacterial activity. Generally speaking, mercury in sediments of water bodies is converted faster under aerobic conditions than in sediments with low oxygen levels or where anaerobic conditions prevail. Methylmercury can readily enter the complex aquatic food chain which may include many levels of concentration.

Mercury compounds, regardless of whether they originate from natural activities or are introduced into the aquatic environment as contaminants, can enter fish via two pathways (Raeder and Snekvik, 1941; Jernelöv, 1972b). The most easily understood path is through the food chain or the food web. All studies on the food chain show a stepwise mercury concentration increase at each subsequent level, culminating in the large predatory organisms, i.e., tunas, billfishes, and sharks. The complexity of the food webs, i.e., number of steps and number of interrelations, and the metabolic requirements of the various members, usually limit the amount of mercury found at each level.

Another pathway, the relative importance of which is poorly understood, is direct extraction of mercury from solution in the aqueous media due to the affinity of mercury ions for proteins. The possibility of the direct absorption of mercury from water was postulated back in 1941 by two Norwegian scientists, Raeder and Snekvik (1941). The most obvious type

of absorption would be through the gills, but it was also found that methylation of inorganic mercury takes place in the slime of some fishes (Jensen and Jernelöv, 1969). The ultimate fate of methylmercury formed this way has not been investigated to the best of our knowledge, but because the organic forms of mercury have an affinity for lipids, perhaps the methylated form also enters the tissues of the fish. It should be pointed out that research on freshwater fishes demonstrates that methylation in fish slime is accomplished mainly, if not exclusively, by certain microorganisms.

Tunas and billfishes, which have high metabolic rates and are extremely mobile species, filter many thousands of liters of water over a short period of time. Thus the possibility of branchial extraction of mercury may be of importance to the buildup of this substance in tunas, especially in regions where the relative concentrations of dissolved mercury are high. The existence of specific oceanic areas with high concentrations of mercury throughout the water column has been demonstrated (Weiss et al., 1972). As food organisms would also tend to have greater mercury content in such regions, it is readily conceivable that a disproportionate increase in mercury levels could be detected in tunas from such regions. Although areas with high volcanic activity would be suspected of having higher amounts of dissolved mercury, there are no data to verify this. Indeed very little is known about geographic variations in concentration of mercury in the world oceans. Furthermore, it is not yet known which of the two pathways responsible for the presence of mercury in tunas is more important. From experiments with other fishes it has been demonstrated that the mercury in some species originates principally from their food whereas in other fishes the mercury originates mainly from mercury dissolved in water (Jernelöv, 1972b).

Research dealing with types of mercury in fishes indicates that in most instances practically all of the mercury accumulated is in an organic form, namely methylmercury (Westöo, 1966, 1967). The same seems to be true of the tunas and swordfish, *Xiphias gladius*. A recent analysis (Kamps, Carr, and Miller, 1972) of the total mercury/methylmercury relation-

ship, based on 11 samples of canned tuna and 20 samples of frozen swordfish steak from the U.S. market, shows that mercury in the edible portions of these fish is essentially all methylmercury. However, it should be noted that Pacific blue marlin, *Makaira mazara*, from Hawaii were found to have total mercury levels ranging from 0.35 to 14.0 ppm<sup>2</sup> whereas the organic mercury for the same samples ranged from 0.23 to 1.79 ppm (Rivers, Pearson, and Shultz, 1972). Furthermore, studies on the methylmercury and total mercury relationship in lake trout, *Salvelinus namaycush*, from Cayuga Lake, N.Y., indicate that 31-35% of total mercury in 1-year fish is methylmercury (Bache, Gutenmann, and Lisk, 1971). By the fourth year, methylmercury in lake trout made up 70% of the total. In 12-year olds, values ranged from 67 to 88%. These findings, as well as some other studies undertaken by a Japanese researcher (Ui, 1971), indicate that the organic to total mercury ratio may not be consistent among or within species and that the relative quantities of the two forms should be further studied.

Not all mercury which enters the body of a fish is retained. For example, the uptake of mercury from food ingested by northern pike, *Esox lucius*, a freshwater fish, does not exceed 20% (Jernelöv, 1972b). Experiments carried out recently by the National Marine Fisheries Service indicate that an even lower figure is applicable to skipjack tuna, *Katsuwonus pelamis*, (Lasker and Leong, 1972).<sup>3</sup> It has been shown that some fish have the ability to rid themselves of some of the mercury which enters the tissues of their body. The mercury excretion rate for tuna is unknown, but for some other species of fish the biological half-life for methylmercury is known to be on the order of 2 yr (Järvenpää, Tillander, and Mietinen, 1970; Lockhart et al., 1972).

<sup>2</sup> Parts per million by wet weight, i.e., 1 mg of metallic mercury per kilogram of wet weight corresponds to 1.0 ppm.

<sup>3</sup> Lasker, R., and R. L. Leong. 1972. Uptake and excretion of mercury by skipjack tuna in aquaria. Southwest Fish. Cent., Natl. Mar. Fish. Serv., NOAA, La Jolla, CA 92037. (Unpubl. manuscr.)

## LEVELS OF MERCURY IN TUNAS AND OTHER LARGE PELAGIC MARINE FISHES

Published data on levels of mercury in tunas and other large pelagic fishes are scarce and difficult to interpret. For example, it has been shown for skipjack tuna caught off Hawaii that the amount of mercury in the red muscle is about 1.6 times greater than that in the white muscle (Lasker and Leong, see footnote 3), and for swordfish caught off the Canadian and U.S. Atlantic coast the amount of mercury in the red muscle is about 1.4 times greater than in the dorsal muscle (Beckett and Freeman, in press), but the majority of published data do not name the parts of the fish which were analyzed.

Table 1 summarizes information on mercury levels in tunas and billfishes, obtained princi-

pally from data published within the last 2 yr. The area designations marked with a 4 or 5 refer to museum specimens all of which date from the period 1878-1901 (except for the swordfish which was captured in 1946) and thus predate the period in which mercury pollution became important. It is evident that there is no significant difference in mercury levels between the museum tuna samples captured 62-93 years ago and the tunas caught within about the last 5 yr. The mercury level from the single museum specimen of swordfish falls within the range of the six "modern" specimens caught off California.

The similarity in mercury levels of tunas collected prior to 1900 and those captured in the last several years is substantiated by similar findings in other types of marine fishes. For example, in a recent study of mercury in benthopelagic fishes it was found that mercury

TABLE 1.—Amounts of total mercury expressed as parts per million (wet weight), in tunas and billfishes.

| Species and area                            | No. of specimens | Weight (kg) | Fork length (cm) | Mercury (ppm)           |       | Reference  |
|---|------------------|-------------|------------------|-------------------------|-------|--|
|   |                  |             |                  | Range                   | Mean  |  |
| <i>Yellowfin tuna, Thunnus albacares:</i>   |                  |             |                  |                         |       |  |
| Gulf of Guinea                              | 88               | 2-105       | —                | 0.07 -1.20              | 0.42  | Ivory Coast Fisheries Service, 1972 <sup>1</sup> |
| Hawaii                                      | 22               | 31-98       | —                | 0.24 -1.32              | 0.54  | Rivers et al., 1972                              |
| Pacific                                     | 6                | 4-37        | —                | 0.20 -0.76              | —     | Unpublished data                                 |
| Atlantic (Africa)                           | 3                | 4.5-40.0    | —                | 0.29 -0.77              | 0.49  | Establier, 1972                                  |
| <i>Skipjack tuna, Katsuwonus pelamis:</i>   |                  |             |                  |                         |       |  |
| Hawaii                                      | 26               | 0.6-1.8     | —                | <sup>2</sup> 0.15 -0.35 | —     | Lasker and Leong, 1972 <sup>3</sup>              |
|   | 20               | —           | —                | 0.27 -0.52              | 0.38  | Rivers et al., 1972                              |
| Gulf of Guinea                              | 5                | 2-6         | —                | 0.11 -0.20              | —     | Ivory Coast Fisheries Service, 1972 <sup>1</sup> |
| Pacific                                     | 1                | —           | —                | —                       | 0.18  | Miller et al., 1972                              |
| Massachusetts <sup>4</sup>                  | 2                | —           | —                | 0.27 -0.64              | 0.46  | Miller et al., 1972                              |
| Philippines <sup>4</sup>                    | 1                | —           | —                | —                       | 0.26  | Miller et al., 1972                              |
| Pacific                                     | 1                | —           | —                | —                       | 0.18  | Miller et al., 1972                              |
| San Diego, Calif. <sup>4</sup>              | 1                | —           | —                | —                       | 0.45  | Miller et al., 1972                              |
| Hawaii                                      | 1                | —           | —                | —                       | 0.42  | Miller et al., 1972                              |
| <i>Albacore, Thunnus alalunga:</i>          |                  |             |                  |                         |       |  |
| California <sup>4</sup>                     | 1                | —           | —                | —                       | 0.27  | Miller et al., 1972                              |
|   | 1                | —           | —                | —                       | 0.13  | Miller et al., 1972                              |
|   | 2                | —           | —                | 0.183-0.209             | 0.193 | Knauer and Martin, pers. comm.                   |
| <i>Bluefin tuna, Thunnus thynnus:</i>       |                  |             |                  |                         |       |  |
| Gulf of Cádiz                               | 6                | —           | 200-271          | 0.46 -0.91              | 0.68  | Establier, 1972                                  |
| Western Atlantic                            | 1                | —           | 172              | —                       | 0.80  | Beckett and Freeman, in press                    |
| Massachusetts <sup>4</sup>                  | 1                | —           | —                | —                       | 0.38  | Miller et al., 1972                              |
| <i>Bigeye tuna, Thunnus obesus.</i>         |                  |             |                  |                         |       |  |
| Gulf of Guinea                              | 5                | 30-70       | —                | 0.23 -0.75              | —     | Ivory Coast Fisheries Service, 1972 <sup>1</sup> |
| <i>Swordfish, Xiphias gladius:</i>          |                  |             |                  |                         |       |  |
| Western Atlantic                            | 210              | —           | 74-247           | 0.05 -4.90              | 1.15  | Beckett and Freeman, in press                    |
| California                                  | 6                | —           | —                | 0.23 -1.27              | —     | Miller et al., 1972                              |
| Gibraltar Strait                            | 5                | —           | —                | 0.99 -2.01              | 1.36  | Establier, 1972                                  |
| Gulf of Guinea                              | 4                | —           | —                | 0.95 -1.25              | —     | Ivory Coast Fisheries Service, 1972 <sup>1</sup> |
| Baja California <sup>5</sup>                | 1                | —           | —                | —                       | 0.52  | Miller et al., 1972                              |
| <i>Pacific blue marlin, Makaira mazara:</i> |                  |             |                  |                         |       |  |
| Hawaii                                      | 27               | 44-355      | —                | 0.35 -14.0              | 4.78  | Rivers et al., 1972                              |
| <i>White marlin, Tetrapturus albidus:</i>   |                  |             |                  |                         |       |  |
| Western Atlantic                            | 1                | —           | 187              | —                       | 1.34  | Beckett and Freeman, in press                    |

<sup>1</sup> See text footnote 4.

<sup>2</sup> White meat only.

<sup>3</sup> See text footnote 3.

<sup>4</sup> Museum specimens collected from 1878 to 1901.

<sup>5</sup> Museum specimen captured in 1946.

levels in specimens taken in 1971-72 do not differ significantly from those in the same species captured in the same area 85-90 yr ago (Barber, Vijayakumar and Cross, 1972). Also, mercury levels in eight fish, between 1605 and 2100 yr old, discovered in Michigan and Illinois archeological sites, were found to be as high as 0.515 ppm; and nine marine fish from a pre-Inca site in Peru contained as much as 9.5 ppm (Medical World News, 1972).

By way of comparison, mercury levels of some other selected marine fishes are shown in Table 2. These data seem to support the general concept that larger predatory fishes, including tunas, have higher mercury levels than those fish species which occupy a lower level in the food chain.

## RELATIONSHIP BETWEEN MERCURY LEVELS AND FISH SIZE

Swedish investigators (Johnels et al., 1967) reported that mercury levels in northern pike increase with the age of the fish. A similar relationship has been reported for freshwater fishes from Wisconsin (Kleinert, 1972).

Mercury content is also known to vary with fish size in some marine species, e.g., in swordfish captured off the Atlantic coast of Canada and the United States (Beckett and Freeman, in press), in Pacific halibut, *Hippoglossus stenolepis*, of the North Pacific coast of Canada and the United States (Bernard E. Skud, International Pacific Halibut Commission, pers. commun.), in a benthopelagic morid *Antimora rostrata* from the U.S. east coast (Barber et al., 1972), in spiny dogfish, *Squalus acanthias*, in the Strait of Georgia, British Columbia (Forrester, Ketchen, and Wong, 1972), and in Pacific blue marlin from Hawaii (Rivers et al., 1972).

A similar relationship has also been demonstrated in some species of tunas. One of the most detailed studies (Ivory Coast Fisheries Service, 1972)<sup>4</sup> is that for 88 yellowfin tuna, *Thunnus albacares*, captured in the Gulf of Guinea between July 1971 and July 1972 (Figure 1). The large amount of scatter in

<sup>4</sup> Ivory Coast Fisheries Service. 1972. La contamination mercurielle des thons. [Mercury contamination in tuna fish.] Ivory Coast Fish. Serv., Abidjan, 5 p. (Engl. transl. by U.S. Embassy, Abidjan, 7 p.)

TABLE 2.—Amounts of total mercury, expressed as parts per million (wet weight), in some selected marine fishes.

| Area                                      | Species  | No. of specimens | Fork length (cm) | Mercury (ppm) |       | Reference                      |
|---|--|------------------|------------------|---------------|-------|--------------------------------|
|   |  |                  |                  | Range         | Mean  |                                |
| Hawaii:                                   |  |                  |                  |               |       |                                |
|   | Squirrel fish, <i>Myripristis arayomus</i>         | 14               | —                | 0.10 -0.43    | 0.21  | Rivers et al., 1972            |
|   | Red goat fish, <i>Mulloidichthys auriflamma</i>    | 10               | —                | —             | <0.05 | Rivers et al., 1972            |
|   | Bige-eyed scad, <i>Trachurops crumenophthalmus</i> | 10               | —                | 0.07 -0.11    | 0.09  | Rivers et al., 1972            |
|   | Mullet, <i>Mugil cephalus</i>                      | 10               | —                | —             | <0.05 | Rivers et al., 1972            |
| Monterey Bay, Calif.:                     |  |                  |                  |               |       |                                |
|   | Myctophid, <i>Tarletonbeania crenularis</i>        | 29               | —                | 0.013-0.096   | 0.030 | Knauer and Martin, 1972        |
|   | Myctophid, <i>Diaphus theta</i>                    | 5                | —                | 0.032-0.078   | 0.060 | Knauer and Martin, 1972        |
|   | Pacific saury, <i>Cotolabis saira</i>              | 2                | —                | 0.008-0.011   | 0.009 | Knauer and Martin, pers. comm. |
|   | Northern anchovy, <i>Engraulis mordax</i>          | 3                | —                | 0.055-0.076   | 0.063 | Knauer and Martin, 1972        |
| Oregon:                                   |  |                  |                  |               |       |                                |
|   | Rex sole, <i>Glyptocephalus zachirus</i>           | 82               | —                | 0.05 -0.24    | 0.119 | Childs and Gaffke, 1973        |
|   | Petrale sole, <i>Eopsetta jordani</i>              | 94               | —                | 0.05 -0.32    | 0.114 | Childs and Gaffke, 1973        |
|   | Starry flounder, <i>Platichthys stellatus</i>      | 42               | —                | 0.08 -0.50    | 0.235 | Childs and Gaffke, 1973        |
|   | Yellowtail rockfish, <i>Sebastes flavidus</i>      | 24               | —                | 0.19 -0.53    | 0.371 | Childs and Gaffke, 1973        |
|   | Rougheye rockfish, <i>Sebastes aleutianus</i>      | 30               | —                | 0.06 -0.11    | 0.080 | Childs and Gaffke, 1973        |
|   | Lingcod, <i>Ophiodon elongatus</i>                 | 48               | —                | 0.06 -0.73    | 0.351 | Childs and Gaffke, 1973        |
|   | Sablefish, <i>Anoplopoma fimbria</i>               | 52               | —                | 0.03 -0.65    | 0.138 | Childs and Gaffke, 1973        |
|   | Pacific hake, <i>Merluccius productus</i>          | 24               | —                | 0.06 -0.18    | 0.102 | Childs and Gaffke, 1973        |
|   | Arrowtooth flounder, <i>Atheresthes stomias</i>    | 26               | —                | 0.01 -0.33    | 0.154 | Childs and Gaffke, 1973        |
|   | Spiny dogfish, <i>Squalus acanthias</i>            | 88               | —                | 0.20 -1.14    | 0.602 | Childs and Gaffke, 1973        |
| British Columbia:                         |  |                  |                  |               |       |                                |
|   | Spiny dogfish, <i>Squalus acanthias</i>            | 206              | 125-120          | 0.1 -1.96     | —     | Forrester and Ketchen, 1972    |
| Atlantic coast, Canada and United States: |  |                  |                  |               |       |                                |
|   | Basking shark, <i>Cetorhinus maximus</i>           | 2                | 382              | 0.03 -0.14    | 0.08  | Beckett and Freeman, in press  |
|   | Blue shark, <i>Prionace glauca</i>                 | 14               | 69-190           | 0.40 -1.17    | 0.70  | Beckett and Freeman, in press  |
|   | Sickle shark, <i>Carcharhinus falcaformis</i>      | 4                | 101-199          | 0.75 -3.28    | 1.43  | Beckett and Freeman, in press  |
|   | Mackerel shark, <i>Lamna nasus</i>                 | 4                | 78-234           | 0.62 -5.43    | 2.08  | Beckett and Freeman, in press  |

<sup>1</sup> Total length.

the points among fish weighing more than about 65 kg is borne out by the following data on white muscle of eight yellowfin tuna taken by a single vessel during one day's fishing in the Gulf of Guinea:

| Fish weight<br>(kg) | Mercury level<br>(ppm) |
|---------------------|------------------------|
| 66                  | 0.78                   |
| 67                  | 0.39                   |
| 70                  | 0.86                   |
| 70                  | 0.29                   |
| 70                  | 0.44                   |
| 75                  | 0.45                   |
| 75                  | 0.19                   |
| 75                  | 0.58                   |

The large variation in mercury levels among tunas of the same size may be due to a differential growth rate between sexes. For example, Forrester et al. (1972) noted that for any given length above 65 cm, the mercury content was higher among male spiny dogfish than among females. They pointed out that since mercury has been shown to be cumulative with age in some species (Johnels et al., 1967) the difference in mercury levels between sexes may reflect differences in growth rates since it is probable that males grow slower than females beyond a certain age. Moreover, large

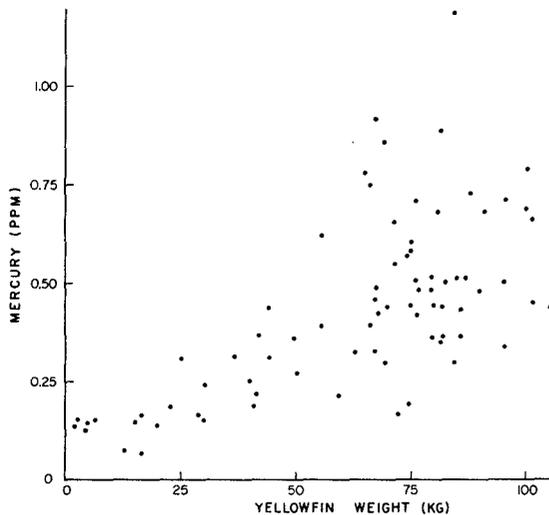


FIGURE 1.—Relation between mercury (ppm) and fish weight (kg) for 88 yellowfin tuna captured in the Gulf of Guinea between July 1971 and July 1972 (from Ivory Coast Fisheries Service, 1972, see footnote 4).

differences in growth rates among individuals of the same sex may further complicate the relationship.

In a study of 22 yellowfin tuna captured off Hawaii (size range 31-98 kg; mercury level 0.24-1.32 ppm) a relationship was found between fish weight and total mercury level ( $r = 0.47$ ;  $P = 0.05$ ) (Rivers et al., 1972).

Also, in a study of 26 Hawaiian skipjack tuna it was found that mercury levels in the white muscle of the smaller specimens (18 fish), weighing from 0.6 to about 1.4 kg, were at a nearly constant level of about 0.15 ppm, whereas mercury in the larger fish (8 specimens), weighing about 1.5 to 1.8 kg, ranged from approximately 0.15 to 0.35 ppm (Lasker and Leong, see footnote 3).

## GEOGRAPHIC VARIATION IN MERCURY CONTENT OF FISHES

Geographic variations in the mercury levels of fishes have been demonstrated in many cases. Some of these variations have been related to mercury pollution by man such as that observed in some Swedish lakes, Minamata Bay in Japan, and some of the Great Lakes and connecting waters between the United States and Canada.

Other cases of geographic variation have been observed, but it is not known whether they are related to human activities. For example, an investigation of north Pacific halibut has shown that there is a south-north cline in mercury levels, i.e., halibut from the southern areas (off Oregon and Washington States) have higher levels than those from the more northern areas. Also it was found that there are localized "hot spots" in the halibut fishery where the fish have significantly higher mercury levels on the average than halibut in the surrounding area (Bernard E. Skud, pers. commun.). Also Childs and Gaffke (1973) found significant geographic variation in mercury levels of some groundfish (rex sole, *Glyptocephalus zachirus*; dover sole, *Microstomus pacificus*; sand sole, *Psettichthys melanostictus*; starry flounder, *Platichthys stellatus*; lingcod, *Ophiodon elongatus*; and sablefish, *Anoplopoma fimbria*) from three areas off the Oregon coast. However it is interesting to note

that this variation was inconsistent among the species examined, e.g., mercury concentration in some species was highest in the northern area while for other species from the same area it was lowest.

A proper understanding of the cause(s) of geographic variations in mercury levels of fishes is confounded by the possibility that a complex of factors may be involved. For example, Beckett and Freeman (in press) in a study of 210 swordfish from six areas extending from the Caribbean Sea to the Grand Banks found that the average mercury level of this species varied significantly from area to area. They suggested that this variation may be related to (1) the passage of time, i.e., the mercury content appeared to decrease with time in the northern part of the area of investigation; (2) a higher rate of mercury uptake and/or excretion in the southern areas; (3) differences in size composition of swordfish among areas; and (4) the occurrence of forage fishes of high mercury content near the Grand Banks.

Furthermore, Forrester et al. (1972), in a study of about 200 spiny dogfish from four areas in the Strait of Georgia, British Columbia, found differences in mercury content of this species among areas. Mercury level was highest in the Point Grey—U.S. Border area. The authors suggest that contamination by effluents from industrial plants along the Fraser River and its estuary may be a contributing factor. They go on to point out that "The differences in mercury content among areas suggest that dogfish in Georgia Strait are not a homogeneous stock or, if they are, that mercury uptake is very rapid." The authors also state that "If uptake is rapid the extreme variation in mercury content for dogfish of a given length may indicate time spent by various fish in the region of apparent high mercury contamination. On the other hand, there may be large differences in growth rates among individuals, reflecting differences in accumulation with time."

Finally, it is interesting to note that mercury levels in *Aldrovandia macrochir*, a benthopelagic fish captured off the U.S. Atlantic coast, were about an order of magnitude lower than those of several other benthopelagic fishes—macrou-

rids and morids (*Antimora rostrata*, *Bathysaurus agassizi*, *Chalinura brevibarbis*, and *C. carapina*)—from the same area-time stratum and of about the same average length, even though their feeding habits and ecological requirements appear to be very similar (Barber et al., 1972). The authors suggest the existence of fundamental metabolic differences to account for the great differences in mercury concentration.

We were unable to find any published data on geographical variations in the mercury levels of tunas, but it is reasonable to suppose that they do occur in view of (1) the geographic variation of mercury concentrations in seawater (Weiss et al., 1972); (2) the geographic exposure of tunas—they are known to occur in all of the major temperate and tropical oceans of the world, and some species migrate many thousands of miles; and (3) the fact that tunas are known to be quite nonselective in their feeding habits (Alverson, 1963).

## MERCURY CONTAMINATION AND TOXICOLOGY

As stated earlier, although mercury has been present in our environment since the beginning of time, there are indications that man has been adding significant amounts of this metal to the atmosphere, land, and waters. An analysis of mercury in ice in Greenland has revealed a substantial increase in deposition since 1940 (Weiss, Koide, and Goldberg, 1971). In Sweden, concentrations of mercury in bird feathers were measured, and it was found that mercury levels were low in museum specimens of fish-eating birds collected from 1840 to 1880-90. In the years following, the level has been continuously on the rise. An analysis of seed-eating birds and birds which prey upon them did not show a corresponding rise in levels of mercury until 1946 (Berg et al., 1966). The increase in mercury in the fish-eating birds has been associated with the period when Sweden started to industrialize whereas the marked increase in the seed-eating birds and terrestrial birds of prey has been related to the introduction of organomercurial dressing for

grain. Since 1965 the mercury levels in the feathers of Swedish seed-eating birds have decreased coinciding with the prohibition of alkyl mercury dressings for seed (Johnels and Westermark, 1969).

It should be noted that the increase of mercury in fish-eating birds in Sweden, based on the examination of museum specimens and those of more recent times, is at variance with the situation noted earlier concerning museum specimens of tunas and swordfish. This seeming contradiction may be due to the fact that these fish species are high-seas animals inhabiting waters which are normally far removed from the concentrated effects of mercury pollution caused by man. Furthermore it has been calculated that if the total amount of mercury processed by man since 1900 were put into the world's oceans and well mixed, it would increase the average mercury concentration of seawater (approximately 0.1 part per billion) by at most 1% (Hammond, 1971). Other investigators (Weiss et al., 1971) have calculated that even if the atmospheric mercury load resulting from man's activities equaled the natural degassing rate (which is very unlikely) it would add annually only 15% to the mercury burden of the upper 100 m of the oceans (the mixed layer). With a residence time of mercury in this layer of as much as 5 yr, the mercury content would be increased by a factor of only 0.75. The increased mercury content in surface waters, if transmitted through the food web to the large predatory fishes such as tunas and billfishes, would, at most, double the mercury content in these animals.

Under circumstances whereby large amounts of mercury enter the aquatic environment, freshwater and marine fishes as well as invertebrates will accumulate large amounts of organic mercury, mostly in the form of methylmercury (Hannerz, 1968). Ingestion of substantial quantities of such contaminated aquatic organisms may lead to toxicological symptoms ranging from temporary or permanent afflictions to death. Fortunately, it appears that the contamination caused by industrial discharges and agricultural use of grain treated with mercury compounds, although persistent, tends to become localized.

## MERCURY POISONING AND ITS EFFECTS

Methylmercury poisoning resulting from ingestion of contaminated aquatic organisms became evident in 1953 when an epidemic of neurological debility and deaths in people and animals which had eaten fish and other seafood from Minamata Bay in Japan was traced to dumping of large amounts of inorganic mercury into the sea and subsequent microbial methylation of that mercury (Ui, 1971).

It is of no surprise that health authorities around the world became concerned about the possible presence of mercury contamination in their waters. In 1967, Sweden became alarmed to the extent that it closed about 40 contaminated freshwater areas to commercial fishing. At the same time Swedish health authorities established a legal limit of 1.0 ppm as the maximum permissible concentration of mercury in the flesh of fish destined for human consumption. In 1970, fishes of the Great Lakes region were investigated and found to have high mercury levels. A survey of fishes in various lakes showed a direct correspondence between mercury levels in fishes and their relative proximity to industrial mercury sources. Fish-eating birds from the same areas were collected for study. Some of the species of aquatic birds were found to have levels of mercury much higher than in the fishes, which has been related to their relative position within the food chain.

When the clinical aspects of mercury poisoning are considered (Katz and Krenkel, 1972) it becomes more evident why such great concern exists in many parts of the world about the occurrence of unduly high amounts of methylmercury in food. All indications are that mercury, in the form of its methyl compound, readily crosses the blood-brain barrier. As with most toxic substances, the effect of methylmercury varies from person to person. Symptoms can manifest themselves gradually so that the afflicted individual scarcely notices, or they can manifest themselves rapidly in agitation, convulsions, or even coma. In extreme cases of poisoning, destruction of the central nervous system can be so rapid that serious,

irreversible damage is done before the cause of the symptoms is diagnosed. Other manifestations of methylmercury poisoning are concentric restriction of the visual field or even blindness, inability to coordinate voluntary muscular movements, and paralysis. As methylmercury readily crosses the blood-placenta barrier, even an unborn child can be poisoned if its mother is exposed to methylmercury poisoning. Very little is known about the subclinical effects of methylmercury.

It is interesting to note that Ganther et al. (1972) found that the increase in mercury content between two lots of canned tuna examined, one of low mercury content (0.32 ppm average) and the other of high mercury content (2.87 ppm), was in an approximate 1:1 molar ratio with the increase in selenium. Furthermore they found that selenium added to the diet of rats in quantities comparable to that found in tuna, decreased methylmercury toxicity in the rats. These authors suggest that selenium in tuna may actually lessen the danger to man of mercury in tuna.

### MERCURY STANDARDS FOR FISH AND FISH PRODUCTS

Currently, only about 13 countries have established standards for the allowable amount of mercury in fish and fishery products for human consumption. These standards, ranging from 0.5 to 1.0 ppm, are expressed in parts per million by wet weight and refer to total metallic mercury (the organic and inorganic compounds of mercury are reduced to metallic mercury during the analysis).

The acceptable limits of mercury in fishes and other aquatic organisms are based on investigations (Berglund et al., 1971) of clinically detectable poisoning of adults sensitive to methylmercury which indicate that manifestations appear at a blood level of  $0.2 \mu\text{g/g}$  (expressed as metallic mercury per gram of whole blood). This level is attained by exposure to 0.3 mg of mercury (as methylmercury) per day taking into account the fact that the human body excretes methylmercury at a rate which eliminates half of the amount present in 70-90 days (Aberg et al., 1969; Berglund et al., 1971).

A prestigious Expert Panel appointed by the Swedish Board of the National Institute of Health, in consultation with the Swedish National Board of Health and Welfare and the Swedish National Veterinary Board, applied a safety factor of 10 to arrive at  $0.02 \mu\text{g/g}$  as an acceptable level of mercury (as methylmercury) in whole human blood and 0.03 mg as an acceptable daily intake of methylmercury (Lambou, 1972).

It is generally accepted by researchers and health authorities that with the exception of contaminated wildlife, grain, etc., food of terrestrial origin is relatively free of mercury in general and of methylmercury specifically, and that the only significant source of methylmercury in the human diet is fish and other aquatic organisms (Nelson et al., 1971). In calculations of dietary intake of methylmercury and in the formulation of mercury standards, generally only this class of food is taken into consideration. Therefore, to evaluate the risk of mercury to the general population it is necessary to know the daily or weekly consumption of fish and other aquatic animals.

The Swedish Commission on Evaluating the Toxicity of Mercury in Fish recommended an allowable weekly intake of mercury equivalent to 210 g of fish containing 1.0 ppm of mercury. However, some experts are cautioning that a level of 1.0 ppm in fish might result in a high mercury intake in some individuals eating more fish than the average for the Swedish population, thus attaining the lowest levels shown clinically to be present in persons sensitive to methylmercury poisoning.

The Swedish findings on mercury contamination of fish from some lakes and rivers caused the Food and Drug Administration (FDA) of the U.S. Government to begin its own evaluation, and an "in-house" standard of 0.5 ppm was adopted in May of 1969. If all fish and shellfish contained 0.5 ppm mercury, the daily limit of 0.03 mg of mercury could be reached by eating 420 g of fish and shellfish per week. A. C. Kolbye of FDA, during a hearing before a Congressional Subcommittee, stated that the average weekly intake of fish in the United States is 280 g. However, a survey of the U.S. Department of Agriculture states that the

average is only 168 g. Regardless of the differences between the two estimates of intake, it is evident that even if we assume that all the fish flesh consumed in the United States has 0.5 ppm of mercury, the average daily intake of methylmercury from that class of dietary items should be below 0.03 mg.

A very recent study (Finch, 1973) based on a computer model indicates that FDA's present interim guideline of 0.5 ppm for mercury in fish could be raised to 1.5 ppm without compromising public safety. The computer model uses the results of a survey of fish consumption patterns of about 1,500 U.S. families, together with known levels of mercury in 52 kinds of fish, to predict the distribution of the daily intakes of mercury from fish, among the survey participants.

In 1970, FDA began to test many fish species for mercury content. In a period of a few months they found that nearly 4% of the canned tuna on the wholesale market contained amounts of mercury in excess of the FDA guideline of 0.5 ppm, ranging up to approximately 1.0 ppm. Species of tuna primarily involved were yellowfin tuna, albacore, *Thunnus alalunga*, bigeye tuna, *T. obesus*, and bluefin tuna, *T. thynnus*. Approximately 12,500,000 standard cans of domestic tuna were voluntarily removed from the U.S. market. At about the same time FDA recalled from the market nearly all swordfish because of high levels of mercury. In May 1971, FDA announced that a 3-mo study showed that all but 42 of 853 samples of swordfish contained mercury at or above the guideline of 0.5 ppm. The U.S. Commissioner of Food and Drugs specifically recommended that the public not eat swordfish.

Although the contamination of fish and aquatic organisms has been clearly demonstrated in a number of countries, including the United States, so far there is no evidence that the mercury in tunas, swordfish, marlins, and some other of the high-seas pelagic fishes can be attributed to contamination resulting from human activities. When FDA introduced the safety guidelines, which eventually were instrumental in removing nearly all swordfish and substantial quantities of canned tuna from the

market, it acted essentially under the assumption that the fish product was "adulterated" by an "added substance." Legal aspects of this action have been thoroughly discussed in an article which appeared in Harvard Law Review (1972).

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NOTE

An important paper on mercury in fishes and shellfishes, including tunas, reached our hands after submission of our manuscript for publication:

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